



Article

Mineral extinction

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Abstract

‘Mineral evolution’ has attracted much attention in the last decade as a counterpart of the long-established biological concept, but is there a corresponding ‘mineral extinction’? We present new geochronological data from uranium-bearing secondary minerals and show that they are relatively recent, irrespective of the age of their primary uranium sources. The secondary species that make up much of the diversity of minerals appear to be ephemeral, and many may have vanished from the geological record without trace. Nevertheless, an ‘extinct’ mineral species can recur when physiochemical conditions are appropriate. This reversibility of ‘extinction’ highlights the limitations of the ‘evolution’ analogy. Mineral occurrence may be time-dependent but does not show the unique contingency between precursor and successor species that is characteristic of biological evolution.

Keywords: mineral evolution, U–Th dating, mineralogy, extinction, mineral ecology, secondary mineral, Archaean

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Introduction

Over the last decade, parallels have been drawn between well-established aspects of evolution and diversity in the biological world and the increase in diversity and complexity of mineral species through the past 4.5 billion years of Earth history. This similarity launched the concept of ‘mineral evolution’, which was initially mooted by Zhabin (1979) and then by Hazen *et al.* (2008). It was subsequently expanded upon in papers such as: Hazen *et al.* (2014), Grew *et al.* (2016), Krivovichev *et al.* (2017) and Grew *et al.* (2019), along with the related idea of ‘mineral ecology’ (Hazen *et al.*, 2015). The generalisation of evolutionary phenomena to non-biological complex systems was discussed in general terms by Hazen and Eldredge (2010). The concept of evolution through geological time in mineralogy has had a significant impact on communication of the science and now underpins displays in major museums such as ‘Die Evolution der Minerale’ gallery at the Naturhistorisches Museum in Vienna, Austria (Koeberl *et al.*, 2018) and The Mignone Halls of Gems and Minerals at the American Museum of Natural History (AMNH, 2017). However, the analogy should be applied with caution, given the fundamental difference in underlying ‘evolutionary’ principles. The analogy arises in part because the living and non-living spheres have interacted and co-evolved (Stewart, 2014). However, we raise the question of how far it can be applied: in particular, whether ‘extinction’ of minerals occurs as it does in biologic systems.

The biological world shows a broad pattern of increasing functional and structural complexity through time, which is shared by minerals despite the absence of genetically-based phylogeny.

Organisms can die out, leaving no progeny, although structurally and functionally convergent species can arise to fill similar ecological niches in other places and times through selection pressures. The explosion in the complexity of life seems more subdued once it is acknowledged that 99.9% of biological species are extinct rather than extant (Raup, 1986; Newman, 1997). The secular diversification of the mineral world may also be modulated by analogous ‘mineral extinction’. We have studied the geochronology of some ‘secondary’ uranium minerals derived by alteration of ‘primary’ uranium source minerals of known age, and found the results to have important implications for the nature of ‘mineral extinction’. The rationale, methods and results of this work are presented and discussed below.

Experimental rationale

The majority of the *ca.* 5500 known mineral species formed near the Earth’s surface, at pressures and temperatures close to ambient relative to the full range of geologically possible conditions. They may form as primary precipitates from liquids and condensates from gases, or through alteration or replacement of other solid phases. In particular, Hazen *et al.* (2008) noted that more than half of known species are hydrated and/or oxidised products formed by reaction of earlier minerals with oxygenated water and that this proportion is even higher for some chemical subclasses: ~80% in the case of uranium minerals. Thus, any count of mineral species at a particular locality or for a particular constituent element is likely to be dominated by such secondary species, and it is meaningful to make a distinction between these and the ‘primary’ minerals whose formation did not involve extensive interaction with the atmosphere and hydrosphere.

For the purposes of the present study, we define ‘secondary’ parageneses to be those where minerals have crystallised after at least partial dissolution of precursors under near-surface

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conditions by fluids with a substantial meteoric-water component. This is consistent with the idea that such minerals tend to be oxidised, as rain and groundwater are sources of oxygen under modern atmospheric conditions. The depths that such fluids can reach thus determine the possible range of pressure–temperature conditions for such secondary mineralisation. The deepest aquifer on Earth, the Great Artesian Basin of Australia, carries meteoric-sourced water down to a depth of 3 km (300 bars lithostatic pressure, close to that of the critical point for water). At a typical geothermal gradient of 25°C/km, this depth corresponds to a temperature of 100°C, but is very far below the boiling point of pure water at that pressure.

Tectonic and atmospheric conditions on Earth have changed through geological time, and it has become evident that this exerts a profound effect on the numbers and abundances of mineral species that have been able to form in different geological periods. This time-dependence has been termed ‘mineral evolution’. The paper by Hazen *et al.* (2008) defined ten mineral-evolutionary stages in the history of the Earth, which correspond to pauses between pulses in the rate of appearance of new mineral species, and can be correlated with changes in the behaviour of the lithosphere, hydrosphere, atmosphere and biosphere. A subsequent series of papers examined this evolutionary behaviour in more detail for minerals of specific chemical elements (e.g. Hazen *et al.*, 2009, 2012; Grew *et al.*, 2016). The change in mineralogical makeup of the crust has controlled age distributions of metallogenesis for various elements (Hazen *et al.*, 2014) and has been associated intimately with biological activity (‘coevolution of life and minerals’) since the Archaean, as shown by the occurrence of stromatolites at 3.5 Ga (Vasconcelos and McKenzie, 2009). There has also been a consistent trend towards mineral species showing a greater range of chemical and structural complexity through time (Krivovichev *et al.*, 2017). One methodological limitation for this type of study is that geochronological data for localities is limited, and data for one mineral of an element have been assumed to apply for all associated minerals of that element, irrespective of their primary or secondary nature. This is immaterial if the secondary species formed relatively soon after the primary minerals and have been preserved with them, but otherwise may overestimate the age of the first appearance for the secondary species, as acknowledged by Hazen *et al.* (2012). The study of boron mineral evolution by Grew *et al.* (2016) showed that the cumulative increase in species and complexity remains in that case, even when blatantly ephemeral recent products are removed from the database for each locality.

Uranium minerals are particularly amenable to geological dating from very small samples, as the predominant isotope ^{238}U is a major constituent of the total U content (97.3 at.%) and radioactive with a half-life comparable to the age of the Earth. Today, uranium occurs in multiple valence states: as relatively immobile U^{4+} in a small number of very insoluble primary minerals, such as uraninite (ideally UO_2); as U^{6+} in a much larger number of species containing the relatively mobile, soluble uranyl cation $[\text{UO}_2]^{2+}$, such as torbernite and francevillite (Fig. 1); and rarely as the intermediate U^{5+} in minerals such as wyartite. The enhanced mobility and ability to form a wide range of mineral structures leads to the great diversity of U^{6+} minerals under near-surface conditions (Burns *et al.*, 1997; Lussier *et al.*, 2016). Thus, uranium mineral diversity has evolved markedly through geological time (Hazen *et al.*, 2009), from the anoxic conditions of the Archaean, dominated by primary and detrital deposits of U^{4+} minerals, through to the present day, when such deposits

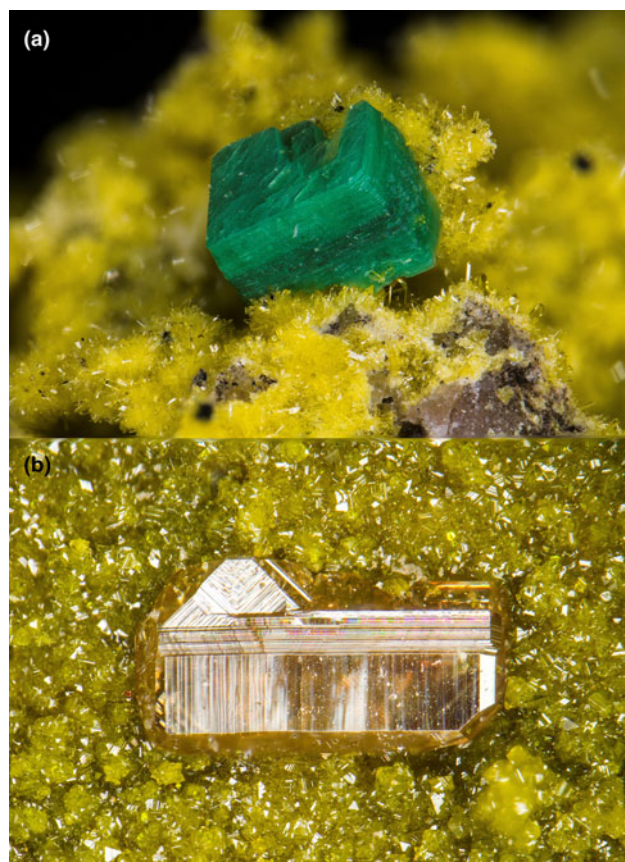


Fig. 1. (a) Torbernite on kasolite from Shinkolobwe, DRC; (b) chervetite on francevillite from Mounana mine, Gabon. Field of view ~ 2.5 mm across. Collection Valérie Galea-Clolus.

oxidise readily to produce a profusion of U^{6+} secondary species. Oxidation also produces dissolved $[\text{UO}_2]^{2+}$, which in turn can be transported and reduced again through contact with organic matter to produce tertiary U^{4+} deposits such as those impregnating sandstones of the Colorado Plateau.

Hazen *et al.* (2009) note that some U^{6+} would have formed through radiation-induced auto-oxidation even in the anoxic Archaean, and that formation of hydrothermal uraninite by reduction from uranyl-bearing fluids became important after the Great Oxidation Event of ~ 2.2 Ga ago, as evidenced by divergence in the geochemical behaviour of U from that of Th, which does not have a mobile hexavalent state. Their deduction that most uranyl minerals only appeared after the Great Oxidation Event seems entirely reasonable. However, ages determined directly from secondary uranyl minerals are scarce in the literature. In this study, we present geochronological data for secondary U minerals from deposits with a wide range of overall age and show that in all cases the secondary minerals are of recent origin, irrespective of the age of the primary U source.

Methods

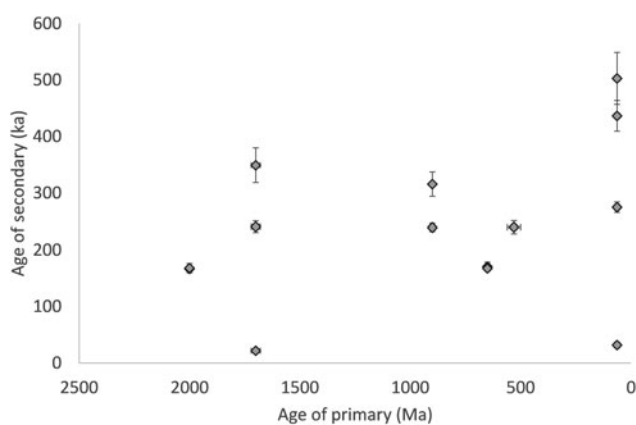
Single crystals were handpicked from specimens in the geological collections of Museums Victoria, Australia (Table 1). The crystals or fragments were weighed into Savillex PFA beakers and dissolved in 1 ml of 7M HNO_3 for 3 days (100°C, capped beaker). On the basis of preliminary tests which indicated high to very

Table 1. U–Th dating samples from a variety of worldwide deposits.

Museum no.	Mineral name*	'Age' (ka)	Error (ka, 2σ)	Locality**	Deposit age	Ref.
M37839	sklodowskite	315.6	21.5	Eva mine, Northern Territory, Australia	~900 Ma	[1]
M36227	parsonsite	240.9	10.1	Ranger U mine, Northern Territory, Australia	~1720 to ~1680 Ma	[1]
M45674	saléeite	21.7	0.3	Ranger U mine, Northern Territory, Australia	~1720 to ~1680 Ma	[1]
M44969	dewindtite	349.1	30.6	Ranger U mine, Northern Territory, Australia	~1720 to ~1680 Ma	[1]
M43448	francevillite	239.5	7.0	Sleisbeck U mine, Northern Territory, Australia	~900 Ma	[1]
M37677	tyuyamunite	239.9	11.3	Musonoi mine, Shaba, DRC	560 to 500 Ma	[2]
M22186	torbernite	170.6	8.0	Shinkolobwe mine, Shaba, DRC	~650 Ma	[3]
M22186	torbernite	167.5	1.9	Shinkolobwe mine, Shaba, DRC	~650 Ma	[3]
M33949	chervetite	166.8	1.7	Mounana mine, Gabon	~2000 Ma	[4]
M37121	mounanaite	168.0	8.2	Mounana mine, Gabon	~2000 Ma	[4]
M52840	moctezumite	436.5	27.1	Moctezuma mine, Sonora, Mexico	~76 to ~50 Ma	[5]
M52841	moctezumite	502.8	46.0	Moctezuma mine, Sonora, Mexico	~76 to ~50 Ma	[5]
M52686	schmitterite	31.9	0.2	Moctezuma mine, Sonora, Mexico	~76 to ~50 Ma	[5]
M52840	schmitterite	274.8	9.1	Moctezuma mine, Sonora, Mexico	~76 to ~50 Ma	[5]

*Formulae: sklodowskite, $\text{Mg}(\text{UO}_2)_2(\text{SiO}_3\text{OH})_2 \cdot 6\text{H}_2\text{O}$; parsonsite, $\text{Pb}_2(\text{UO}_2)(\text{PO}_4)_2$; saléeite, $\text{Mg}(\text{UO}_2)_2(\text{PO}_4)_2 \cdot 10\text{H}_2\text{O}$; dewindtite, $\text{H}_2\text{Pb}_2(\text{UO}_2)_2(\text{PO}_4)_4\text{O}_4 \cdot 12\text{H}_2\text{O}$; tyuyamunite, $\text{Ca}(\text{UO}_2)_2(\text{VO}_4)_2 \cdot 5\text{H}_2\text{O}$; francevillite, $\text{Ba}(\text{UO}_2)_2(\text{VO}_4)_2 \cdot 5\text{H}_2\text{O}$; torbernite, $\text{Cu}(\text{UO}_2)_2(\text{PO}_4)_2 \cdot 12\text{H}_2\text{O}$; chervetite, $\text{Pb}_2(\text{V}_2\text{O}_7)$; mounanaite, $\text{PbFe}_2(\text{VO}_4)_2(\text{OH},\text{F})_2$; moctezumite, $\text{Pb}(\text{UO}_2)(\text{TeO}_3)_2$; schmitterite, $(\text{UO}_2)(\text{TeO}_3)$.
 **Lat/Long: Eva mine: 17°40'48"S, 137°48'36"E; Ranger mine: 12°41'S, 132°55'E; Musonoi mine: 10°43'37"S, 25°27'10"E; Sleisbeck mine: 13°46'51"S, 132°49'36"E; Shinkolobwe mine: 11°2'54"S, 26°33'2"E; Mounana mine: 1°23'54"S, 13°09'09"E; Moctezuma mine: 29°48'N, 109°40'W.

References: [1] Hills and Richards (1976); [2] Porada and Berhorst (2000); [3] Decrée *et al.* (2011); Dewaele *et al.* (2006); [4] Bonhomme *et al.* (1982); [5] McDowell *et al.* (2001).

**Fig. 2.** 'Age' of secondary mineral samples from a variety of worldwide deposits.

high U concentrations and/or high U/Th, small splits of each solution were mixed and equilibrated with a ^{233}U – ^{229}Th tracer. Given the very small sample sizes, these mixtures could be analysed in the mass spectrometer without further matrix removal by ion exchange. Isotopic analyses were carried out on a Nu Plasma multi-collector ICP-MS at the University of Melbourne, using a parallel ion-counting method which measures $^{234}\text{U}/^{238}\text{U}$ and $^{230}\text{Th}/^{238}\text{U}$ activity ratios in a single mass-spectrometer run, with full internal calibration of ion counter gains and mass bias (Hellstrom, 2003; Drysdale *et al.*, 2012). A solution of Harwell uraninite (HU-1), assumed to be in secular equilibrium (McCulloch and Esat, 2000), was used to monitor instrument performance and drift, and provide an estimate of external precision. Further details are given in Supplementary Table S1.

Discussion

We studied samples from a range of geological provinces in the world including those in the Northern Territory, Australia, where the uraninite ages are ~1700 and ~900 Ma; Shaba, Democratic Republic of Congo (560–500 and 650 Ma); the Moctezuma mines in Sonora, Mexico (76–50 Ma); and the natural fission reactor in Gabon (~2 Ga). While the ages determined for

the secondary minerals differ (Table 1), they are geologically young for all host deposits (< 503 ka; Table 1). These results are consistent with those we obtained in Birch *et al.* (2011) for samples of saléeite, ulrichite, torbernite and metanatroautunite from the Lake Boga granite (367 Ma, Mills *et al.*, 2008) and Wycheproof granite (410 Ma) in Victoria, Australia. At those localities, 77 of the 104 samples gave apparent ages <500 ka, with a broad peak of ~400 ka (Marine Isotope Stage 11).

We note that a few studies have found pre-Holocene secondary uranium minerals, but none older than late Miocene. Dill *et al.* (2010) have reported torbernite up to 8.4 Ma occurring in the Bohemian massif of Germany and 1.61 Ma in Galicia, Spain (Dill *et al.*, 2011). These ages are still much younger than their primary U sources in Variscan granite.

The young ages determined from all of the uranyl minerals that we have examined to date emphasises the fact that these are relatively soluble, reactive species that are easily redissolved over long periods of geological time (Fig. 2). While not ephemeral on the scale of years like many evaporite species, they can be regarded as such on the scale of millions of years. Secondary minerals that formed 2.2 Ga ago will not have been preserved intact through that time, except in the rare instances where they are protected as inclusions in some more refractory phase (e.g. Grew, 2017). The same is true for any oxidised secondary minerals that may have formed earlier. The hydrosphere and atmosphere appear to have transitioned to a consistently oxygenated state only after a few hundred million years of oscillations (Koehler *et al.*, 2018), and there is evidence for locally oxygenated microenvironments as early as 3.46 Ga (Hoashi *et al.*, 2009). Photosynthetic production of oxygen long predated the final catastrophic shift in paradigm, which may have been suppressed while excess reducing species were able to scavenge free oxygen effectively (Catling and Claire, 2005). This is the complementary situation to the present day, where despite the prevalent high oxygen fugacity at the Earth's surface, highly reducing environments still occur but are limited in spatiotemporal extent (Konn *et al.*, 2015).

Secondary minerals may persist for long periods if sufficiently abundant or insoluble. Hematite-stained calcare horizons define exhumed and cross-sectioned palaeolandscapes of early Palaeozoic age (>350 Ma) on the island of Arran, Scotland (Friend *et al.*, 1970; Young and Caldwell, 2009). Palaeomagnetic data indicate a

similar age for some Australian regolith environments (Pillars *et al.*, 2004), while other data shows that pisolitic and fragmental duricrust have (U–Th)/He ages of 5.8–1.3 Ma (Wells *et al.*, 2018). Highly insoluble Mn⁴⁺ oxide minerals have yielded ages as old as 65 Ma (Vasconcelos *et al.*, 2013). However, most secondary minerals are scarce in quantity and distribution as well as softer than such resistant oxides, and will have crystallised, only to be eradicated later without trace through chemical attack or mechanical erosion.

Such disappearance would be the fate of the very different suite of secondary minerals that would have formed in the anoxic conditions of the early Archaean. These may well have included highly reduced species, which are likely to have been far more common in surficial environments in the Archaean than their rare and ephemeral modern counterparts.

While occurrences of species can certainly be destroyed, it is in fact very difficult to find examples which can be excluded from recurrence elsewhere or in the future. Even when a particular environment is unlikely to recur on Earth, the spatial domain of mineralogy includes other planetary bodies in this Solar System and beyond, in a way that terrestrial biology cannot. High-temperature, Mg-rich komatiitic lavas, and hence any minerals specific to that environment, are almost confined to the Archaean as a result of secular decrease in geothermal gradients. However, a unique much younger (Paleogene) example of a terrestrial komatiite is known (Gansser *et al.*, 1979) and they erupt still on Io, one of the moons of Jupiter (cf. Williams *et al.*, 2000). An even more unusual ancient rock type, the unique Bon Accord nickel oxide–silicate body, is of Archaean age and uncertain paragenesis (Tredoux *et al.*, 2013; O'Driscoll *et al.*, 2014). However, the only two mineral species that are currently found only in this deposit are the relatively oxidised borate bonaccordite, Ni₂Fe³⁺[BO₃]O₂ (De Waal *et al.*, 1974) and the antimonate tredouxite, NiSb⁵⁺O₆ (Bindi *et al.*, 2018), which could have been formed by boron metasomatism and oxidation of a Ni–Sb sulfide body of any age. Rarity of the physicochemical circumstances required to form a species, and occurrence at only one ancient locality, is likely to provide at most a very temporary form of 'extinct' status.

Given that the species that actually occur are only a subset of those that could do so plausibly (Hazen *et al.*, 2015), there may be many mineral species once present on Earth that are now 'extinct'. Our knowledge of the Earth's former mineralogical inventory is necessarily incomplete, and the apparent secular increase in diversity and complexity may be exaggerated by the loss of species that have long since disappeared. However, while 'virtually all plant and animal species that have ever lived on the earth are extinct' (Raup, 1986), long-absent mineral species may 'return' in the future if favourable conditions recur. This difference in the nature of 'extinction' spotlights the fundamental flaw in the 'evolution' analogy. Phylogenesis of organisms produces unique occurrences of species that are contingent upon the nature of their equally unique predecessors, while paragenesis of minerals lacks this unidirectional, irreversible flow.

Supplementary material. To view supplementary material for this article, please visit <https://doi.org/10.1180/mgm.2019.60>

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